Photocatalytic efficiency of green synthesized ZnO nanoparticles for the degradation of methyl orange dye: A review

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Abstract: The primary global source of water pollution is industrial and textile dyes. These industries produce Highly stable organic dyes that are released untreated into nearby ponds, lakes and rivers. As ZnO nanoparticles (NPs) are inexpensive, non-toxic, chemically and thermally stable, and have a higher UV absorption capacity, they are an effective nano-catalyst for removing hazardous contaminants such as organic and inorganic dyes from wastewater. The green synthesis of synthesised ZnO NPs, their shape and size, and their photocatalytic effectiveness against the dye methyl orange are all described in this paper. The emphasis is on environmentally friendly synthesis of ZnO NPs using plant extracts because the synthesis process impacts the characteristics and applications of ZnO NPs. Additionally, based on recently published literature, the shape and size factors that affect the photocatalytic activity in the degradation of MO dye are highlighted..

Keywords: Green synthesis; Nanoparticles; Photocatalytic activity; Zinc oxide.

1. INTRODUCTION

Nanoparticles (NPs) have become a prominent nanomaterial in today's growing world due to their existing chemical, physical and biological properties (Perveen *et al.*, 2020; Balkrishna *et al.*, 2021a; Kumar *et al.*, 2023a). NPs are tiny particles ranging from 1 to 100 nm in one dimension made from metals, metal oxides, carbon, or other organic materials. They exhibit a unique property at nanoscale due to its relatively large surface area to volume, chemically stability, high reactivity, intensive mechanical strength, etc., (Thi *et al.*, 2020; Anu *et al.*, 2020; Anu, Thakur & Kumar, 2018; Rana *et al.*, 2021). The size and shape NPs vary from different dimensions, as shown in Figure 1. In zero dimensional (0D), nanodots have a single fixed length, width, and height, one dimensional (1D) holds only on a parameter, i.e., graphene, two dimensional (2D) possess length and breadth, i.e., nanotubes and three dimensional (3D) that have all three parameters length, breadth and height, i.e., gold NPs. The NPs consist of unlike size and shape with uniform or irregular structures such as conical, spiral, flat, hollow core, cylindrical, tubular, spherical, etc., (Chang *et al.*, 2020; Bra *et al.*, 2023; Kumar, Chandel & Thakur, 2022a; Patial & Thakur, 2018).

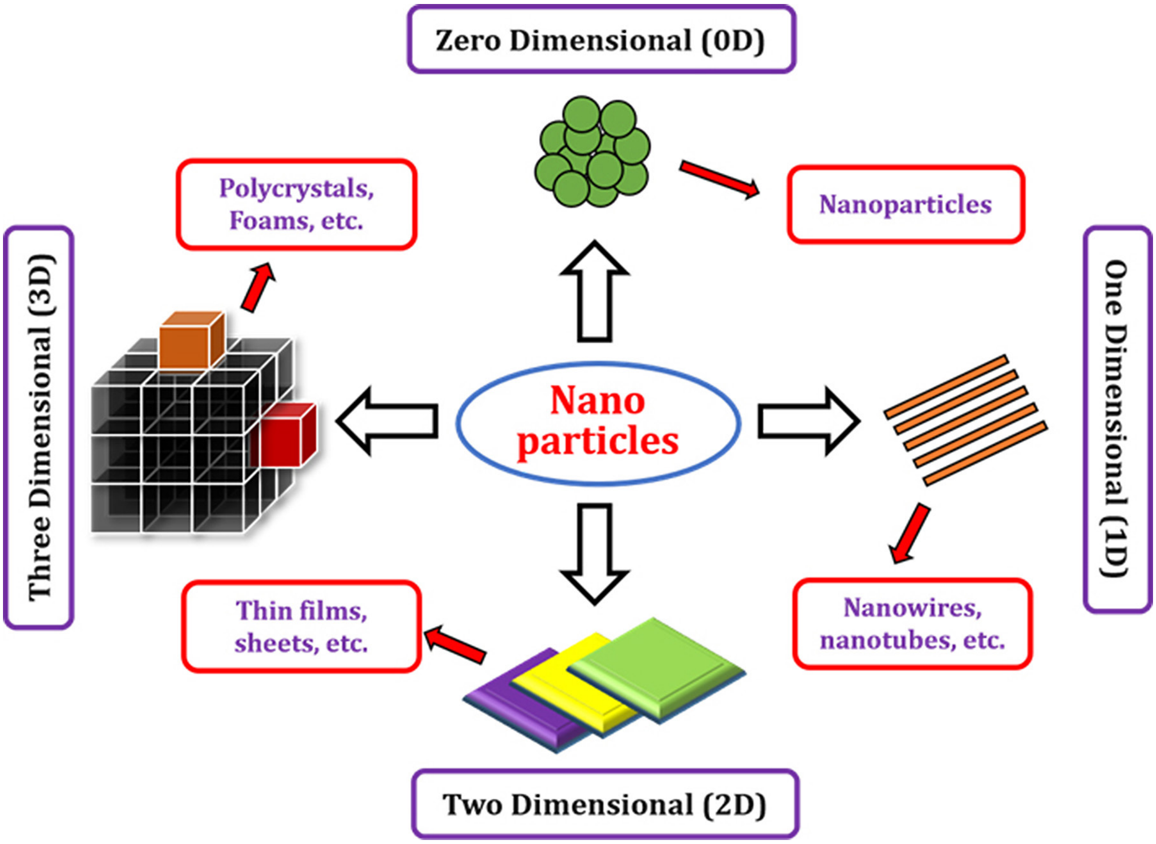

Figure 1. Different dimensions of nanomaterials.

With the success of contemporary companies, particularly those that deal with textile dyes, a significant problem with the acquisition of fresh water has arisen due to the massive amounts of wastewater and other types of effluents that are emitted (Yadav *et al.*, 2021; Kumar *et al.*, 2023b; Kumar *et al.*, 2022b). Pollution of organic dyes such as methyl orange (MO) is a major issues which the world is facing today, include the scarcity of reliable and pure natural energy and environmental hazards (Pal *et al.*, 2018; Sharma, Kumar & Thakur, 2021b). The ecological equilibrium can be easily offended and destroyed by these polluting dyes, which will have a negative impact on all living things, including people and plants. Therefore, there is a pressing need to develop some useful procedures that can turn dangerous and deadly pollutants into harmless ones (Chauhan *et al.*, 2020; Sharma & Kumar *et al.*, 2021a). MO is azo dye utilised in the pigment industry as well as the dyestuff industries because it has ability to turn both alkaline and neutral water in yellow color. Also, in acidic medium, MO dye immediately become red and the transition takes place at the value of pH 4.3. MO has molecular weight of 327.33 gmol^{-1} with molecular formula $C_{14}H_{14}N_3N_aO_3S$ (Prasad *et al.*, 2020) as shown in Figure 2. The chemical structure of MO consists of a benzene ring (C_6H_5) attached to a nitrogen atom (N) and an azo group (-N=N-) through which it is connected to another benzene ring. Additionally, a sulfonate group ($-SO_3^-$) and a sodium ion (Na^+) are attached to the other benzene ring. The presence of the sulfonate group imparts water solubility to MO, making it easily dissolvable in aqueous solutions. The azo group is responsible for the characteristic orange color of MO, which changes to a pinkish-red color in acidic conditions and yellow in basic conditions.

The influence of nano-photocatalysts is recognised as the best for addressing the energy problem, while dye treatment nano-photocatalysts are seen as the most successful at controlling environmental damage issues (Biswal *et al.*, 2021; Thakur *et al.*, 2022a; Thakur *et al.*, 2023a). These nano-catalysts has good conversion capabilities from photons energy to natural energy, which is advantageous

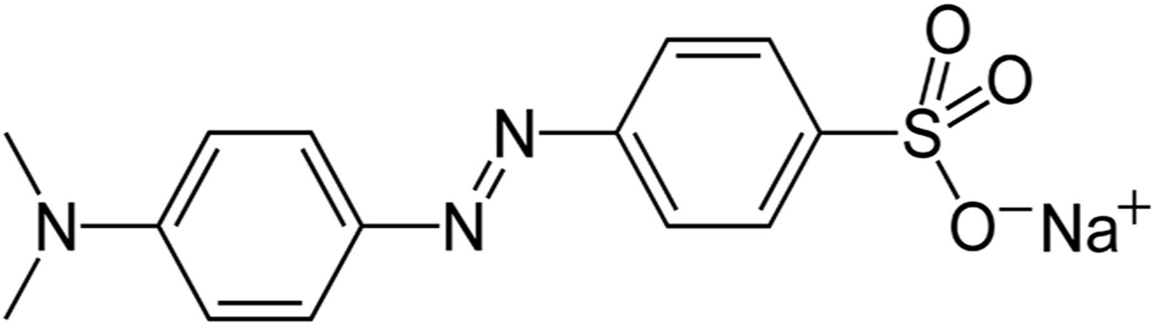

Figure 2. Chemical structure of MO dye.

for the breakdown of the principal hazardous organic pollutants (Jawale *et al.*, 2021; Thakur *et al.*, 2021b; Thakur *et al.*, 2023b). NPs produced through the green synthesis have distinct physiochemical characteristics due to their higher surface area per volume as compared to bulk materials with the same composition (Kumar *et al.*, 2020a). It also allows to use as catalysis for degradation of organic dyes, antimicrobial, anticancer, drug-delivery and several applications (Chakraborty *et al.*, 2020; Sharma *et al.*, 2021c; Thakur *et al.*, 2022b). The green synthesis can manage the size and shape of NPs like nanoporous, nanospheres, nanowires and nanorods which plays an important role in a degradation of various organic dyes (Ansari *et al.*, 2020). Zinc Oxide (ZnO) has been demonstrated to be the most effective potent oxidizing and catalyst agent due to its initial application in heterogeneous photocatalysis under UV light (Soares *et al.*, 2020; Kumar, Thakur & Chandel, 2020b). ZnO has wide bandgap (3.37 eV), high binding energy (60 meV) and deep absorption of ultraviolet (UV) at ambient temperature. Also, it is superior oxide with good electrical, mechanical and optical qualities which is recommended to use in heterogenous photocatalysis (Umavathi *et al.*, 2021; Khatana *et al.*, 2021; Naseer *et al.*, 2020). Heterogeneous photocatalytic oxidation involves a series of stages that are crucial for the successful removal of organic contaminants from a liquid phase. The first stage involves the infiltration of these contaminants onto the surface of ZnO from the liquid phase. Once on the surface, the organic contaminants are absorbed by the ZnO. During the process of absorption, reduction and oxidation processes occur, which further aid in the removal of the contaminants. Once the products are formed, there is a process of desorption that occurs, where the products are separated from the ZnO. Finally, the products are removed from the area of interaction, completing the process of heterogeneous photocatalytic oxidation. These stages are important for understanding the mechanisms involved in photocatalytic oxidation and can help in the development of efficient processes for removing organic contaminants from a liquid phase (Ong *et al.*, 2018).

In this review paper, considering the ZnO as a promising catalyst special focus has given to the shape and size of green synthesized ZnO NPs for the degradation of MO dye using various extract of plants as a capping agent. Additionally, according to studies in the current literature, the degradation efficiency of MO dye utilising ZnO NPs as a photocatalysts is influenced by several parameters that are overviewed in depth. To my knowledge, no previous review of this kind has been done.

2. GREEN SYNTHESIS OF ZnO NPs

Green synthesize ZnO NPs are crucial in the progression of nanostructures from 0D to 3D (Thi *et al.*, 2020; Thakur *et al.*, 2021a). The combination of ZnO NPs with regulated shape and size has different features and applications that apply to biological fields (Umavathi *et al.*, 2021; Thakur, Anu & Kumar, 2020). With the control of size and shape of ZnO, NPs can be easily changed in size and shape, from nanometres to molecules or from spherical to nanorods, nanotubes and nanowires (Zhu *et al.*, 2021; Kumar *et al.*, 2021; Arumugam *et al.*, 2021). In terms of structural phases, controlled morphogenesis and particle size for diverse technical and biosafety applications, the synthesis of ZnO NPs is particularly stimulating (Perveen *et al.*, 2020; Kumar, Chandel & Thakur, 2022a). The green synthesis utilising plant extract is favourable as compared to chemical synthesis, NPs may be created rapidly with minimal precursor costs and low consumption of energy while certifying simpler synthesis procedures (Ahmed *et al.*, 2021; Rao *et al.*, 2021). The upside of employing plant extracts for green synthesis over chemical methods is that it is easier to control shape and size of ZnO NPs. A multicomponent mixture obtained using a suitable solvent during an extraction method might be referred to as a plant extract (Aldeen *et al.*, 2022). Primary and secondary metabolites can generally be used to classify substances produced by plants. Alkaloids, terpenoids, phenolics and other secondary metabolites are examples of those that do not contribute to the growth, development, or reproduction of the plant (Sivasankarapillai *et al.*, 2022). Nucleic acids, carbohydrates, chlorophyll, and other compounds that are involved in the growth of the plant which are examples of primary metabolites. Therefore, extraction involves separating the soluble metabolites from the insoluble plant matrix using an appropriate extraction solvent, frequently referred to as menstruum. (Aldeen *et al.*, 2022; Balkrishna *et al.*, 2021b). Several plant extraction methods, including traditional methods, are available. Secondary metabolites present in extract serve as a capping agent during synthesizing the ZnO NPs and the method used to prepare the extract has an impact on the molecule's composition and concentration (Chandrasekaran *et al.*, 2022; Balkrishna *et al.*, 2021b). Abomuti *et al.* (2021) has synthesized the ZnO NPs using *salvia officinalis* leaf extract having average particle size of 26 nm with hexagonal shape in his research paper as shown in Figure 3. Also, the various plant extract used in synthesizing the ZnO NPs are reported in Table 1 with their shape and size.

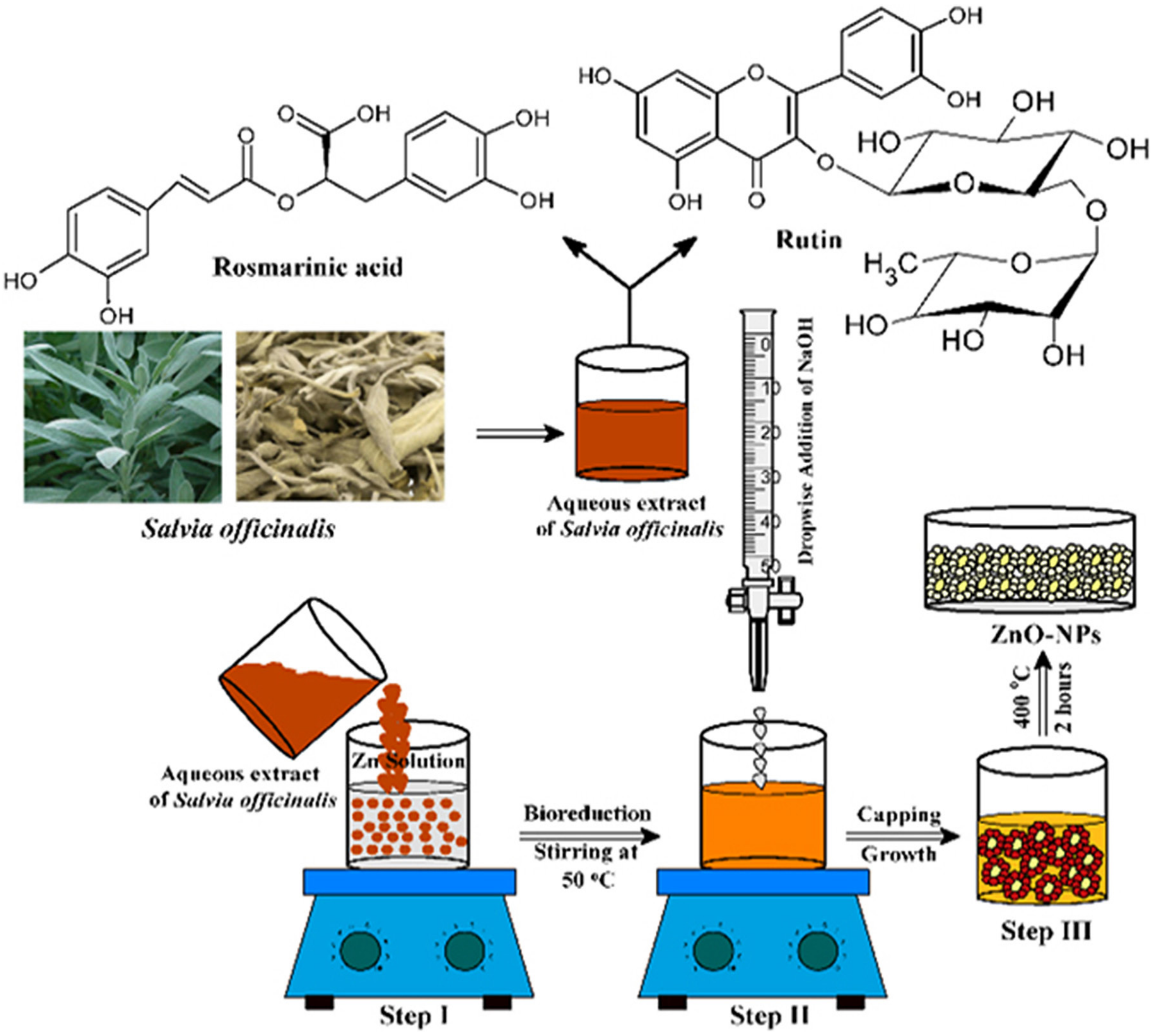

Figure 3. Green synthesis of ZnO NPs by using *salvia officinalis* leaf extract (Reproduced from Abomuti *et al.*, 2021 under the Creative Commons Attribution (CC BY) license, http://creativecommons.org/licenses/by/4.0/).

Method	Precursor	Plant	Shape	Size (nm)	References
Co-precipitation	Zinc acetate dihydrate	*Prunus cerasifera*	Spherical	72-100	Ahmad and Jaffri, 2018
Precipitation	Zinc acetate dihydrate,	*Moringa Oleifera*	Spherical	52	Pal *et al.*, 2018
Precipitation	Zinc acetate	*Cannabis sativa*	Spherical	34	Chauhan *et al.*, 2020
Co-precipitation	Zinc acetate	*Cannabis sativa*	Spherical	38	Chauhan *et al.*, 2020
Precipitation	Zinc acetate dihydrate	*Ocimum tenuiflorum*	Nanorods	30-130	Sharma *et al.*, 2020a
Precipitation	Zinc acetate dihydrate	*Aloe vera*	Spherical	60-180	Sharma *et al.*, 2020b
Precipitation	Zinc acetate	*Gliricidia sepium*	Spherical	56	Prasad *et al.*, 2020
Co-precipitation	Zinc acetate	*Hedyotis capitellata*	Spherical	2-6	Nguyen-Hong *et al.*, 2021
Precipitation	Zinc nitrate	*Salvia officinalis*	Hexagonal	26	Abomuti *et al.*, 2021
Precipitation	Zinc acetate	*Camellia sinensis*	Spherical	10-20	Rao *et al.*, 2021
Precipitation	Zinc nitrate hexahydrate	*Emilia sonchifolia*	Spherical	18-26	D'Souza *et al.*, 2021
Precipitation	Zinc acetate dihydrate	*Urtica dioica*	Spherical	45	Ebrahimian *et al.*, 2021
Precipitation	Zinc chloride	*Livistona jekinsiana*	Spherical	4	Baruah *et al.*, 2021

Table 1. Method, precursor and plant extract used in synthesizing the ZnO NPs.

3. PHOTOCATALYTIC ACTIVITY

The heterogeneous photocatalytic process includes the induced catalytic process, which further oxidises or degrades the organic molecule via a redox reaction (Chauhan *et al.*, 2020). Energy levels over this bandgap will drive electrons (e^-) to the conduction band while leaving h^+ in the top of the valence band to form electron-hole pairs in ZnO, which has a prohibited bandgap of 3.10-3.37 eV (Nguyen-Hong *et al.*, 2021). The space-charge layer successfully separates the photogenerated electrons and holes, with the holes being transported to the surface of the ZnO. The primary photon energy-derived elements that interact with the H_2O or OH^- adsorbed on the surface to make $^\bullet OH$ through vigorous oxidation (Weldegebrieal *et al.*, 2020) as shown in following equations:

$$ZnO + h\nu \rightarrow ZnO\ (h^+ + e^-)$$
$$Dye + h\nu \rightarrow Dye^\bullet$$
$$e^- + O_2 \rightarrow O_2^-$$
$$Dye^\bullet + O_2^- \rightarrow O_2^- + Dye^{+\bullet}$$
$$H_2O + h^+ \rightarrow OH^- + H^+$$
$$Dye + h^+ \rightarrow Dye^+$$
$$Dye + OH^- \text{ or } O_2^- \rightarrow \text{Degraded products}$$

Active free radicals (OH^-, O_2^- and H^+) are generated in this reaction with strong oxidizing properties. By taking use of their powerful oxidizing characteristics, these radicals can immediately mineralize contaminants (mostly organic pollutants) into small inorganic molecules like CO_2 and H_2O and then they can combine with a water molecule to cause the breakdown of MO dye, as shown in Figure 4 (Yaqoob *et al.*, 2020).

Shape and size of NPs can influence the photocatalytic activity towards degradation of various organic dyes. As size of NPs decreases, degradation of dye is expected to increase due to increase in rate of transfer of charge carrier with availability of more active sites. Recombination rate of charge carrier can offset degradation if particle size is too small because of the greater activity brought on by a higher specific surface area (Kusiak-Nejman *et al.*, 2021; sharma *et al.*, 2020b). Abomuti *et al.* (2021)

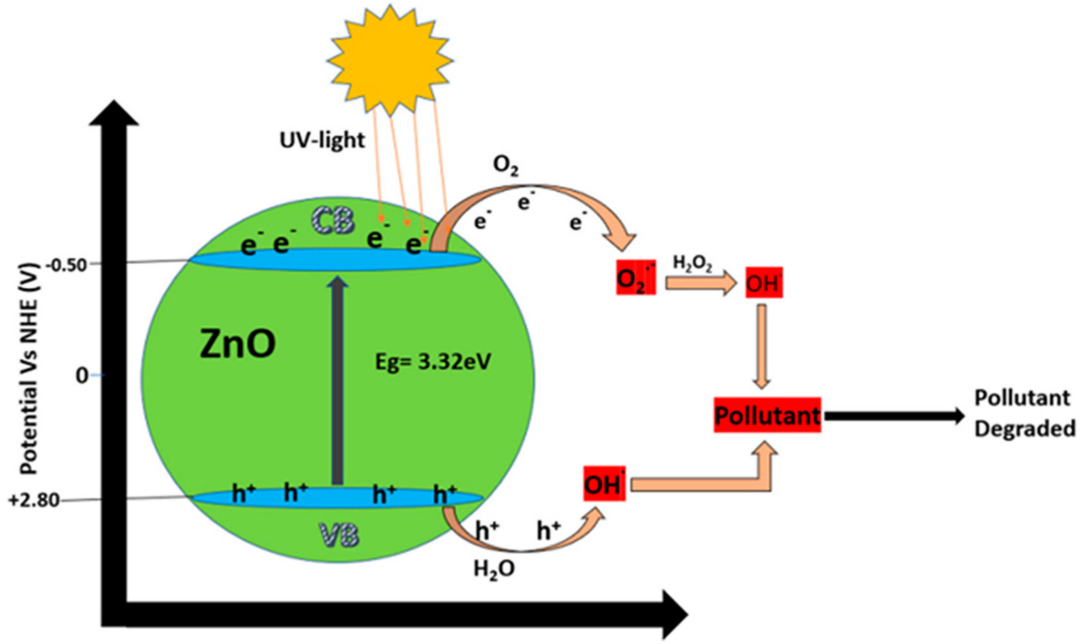

Figure 4. Mechanism of photocatalytic activity of ZnO NPs for degradation of pollutant dyes (Reproduced from Yaqoob *et al.,* 2020 under the Creative Commons Attribution (CC BY) license, http://creativecommons.org/licenses/by/4.0/)

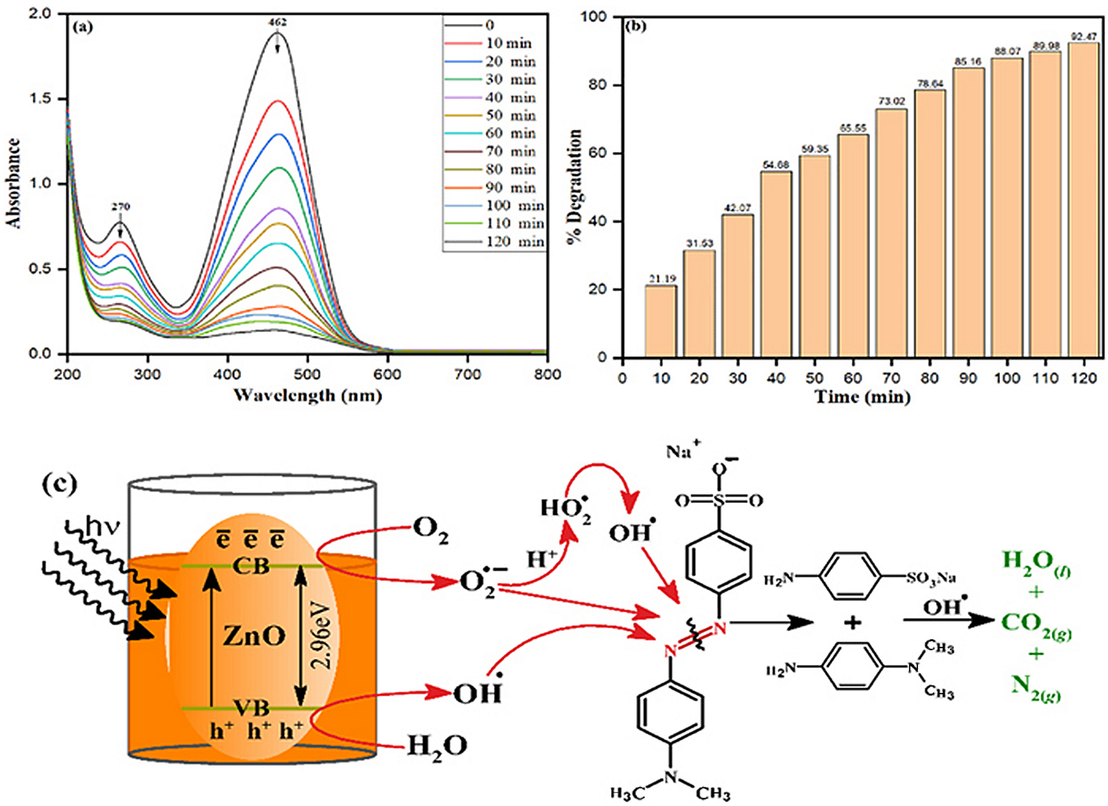

Figure 5. Degradation graph of MO dye by capping plant extract of *salvia officinalis* in ZnO NPs with mechanism (Reproduced from Abomuti *et al.,* 2021 under the Creative Commons Attribution (CC BY) license, http://creativecommons.org/licenses/by/4.0/)

has degraded the MO dye by capping plant extract of *salvia officinalis* in ZnO NPs. The degradation MO dye was recorded upto 92 % in 120 minutes under the exposure of UV light. Also, the degradation of MO dye by using various plant capped ZnO NPs are reported in Table 2 and the plant extract used in synthesis ZnO NPs with their shape and size are already discussed in Table 1.

Source used for photocatalytic activity	Concentration of NPs used for degradation (mg)	Shape	Size (nm)	Degrade time (min)	% Degradation	References
UV Lamp	50	Spherical	72-100	20	82	Ahmad and Jaffri, 2018
Sun light	100	Spherical	52	50	96	Pal *et al.*, 2018
Solar light	—	Spherical	34	80	35	Chauhan *et al.*,2020
Solar light	—	Spherical	38	80	94	Chauhan *et al.*,2020
UV light	25; 50; 100	Nanorods	30-130	180	66; 88; 91	Sharma *et al.*,2020a
UV light	25; 50; 100	Spherical	60-180	160	68; 86; 94	Sharma *et al.*,2020b
UV light	50	Spherical	56	50	93	Prasad *et al.*,2020
UV light	50	Spherical	2-6	240	92	Nguyen-Hong *et al.*,2021
UV light	40	Hexagonal	26	120	92	Abomuti *et al.*,2021
UV light	500	Spherical	10-20	180	80	Rao *et al.*,2021
UV light	50	Spherical	18-26	180	92	D'Souza *et al.*,2021
UV light	100	Spherical	45	90	74	Ebrahimian *et al.*,2021
Sun light	2.5	Spherical	4	60	77	Baruah *et al.*,2021

Table 2. Degradation of MO dye using plant capped ZnO NPs

4. CONCLUSIONS

This review paper discusses research studies on the use of green synthesized ZnO NPs as a photocatalyst for the degradation of persistent organic pollutants, particularly MO dye. The size and shape of ZnO NPs can be controlled using plant extracts as a capping agent, which has been shown to enhance their efficacy in degrading organic dyes. The study found that smaller-sized and spherical-shaped green synthesized ZnO NPs demonstrated the highest efficiency in degrading MO dye. Therefore, green synthesized ZnO NPs can serve as a cost-effective and environmentally friendly solution for the degradation of organic dyes released into water bodies by industries. This method is energy-efficient, fast, and non-toxic to the environment. Thus, the use of plant-capped ZnO NPs can be recommended to industries for large-scale synthesis of nanoparticles to remove specific dyes from wastewater.

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Conflict of interest

The authors declare no competing financial interest.

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